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**NUCLEAR SAFETY IN CHEMICAL  
AND METALLURGICAL PROCESSING  
OF PLUTONIUM**

**E. D. CLAYTON**

**APRIL, 1961**

**HANFORD LABORATORIES**

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NUCLEAR SAFETY IN CHEMICAL  
AND METALLURGICAL PROCESSING OF PLUTONIUM

By

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Nuclear Physics  
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ABSTRACT

A review is made of those types of criticality problems encountered in a typical plutonium processing and metal fabrication plant. A brief discussion is given of some of those criticality data which are of general interest in nuclear safety application, and of some of those data of limited application, but which are of special interest to specific processes. Curves are presented, based on multigroup diffusion theory, which show the estimated critical mass and infinite cylinder diameters for homogeneous  $\text{PuO}_2$ -water mixtures and the critical mass for Pu-Al alloy. Applied methods of criticality control in plutonium processing and fuel element fabrication are reviewed. A list of typical administrative procedures, which have been used in effecting criticality control, is given. The proposed general program of studies for the new Hanford Plutonium Critical Mass Laboratory, which is being undertaken to obtain needed criticality data for Pu solutions and precipitates of plutonium, is discussed.

## NUCLEAR SAFETY IN CHEMICAL AND METALLURGICAL PROCESSING OF PLUTONIUM

### INTRODUCTION

When fissile materials are handled in significant quantities, such as in chemical separations plants for reprocessing spent reactor fuels, and concomitant metallurgical processes, a potential criticality hazard exists.

The prevention of a criticality incident, or inadvertent uncontrolled critical nuclear reaction, is of primordial importance because of the potentially serious consequences which can result.

The nuclear safety problems which are encountered in the handling and processing of fissile materials outside of reactors are especially complex; not only must criticality be considered for the standard operating conditions of the plant, but also for those off-standard conditions which are physically possible and at the same time, perhaps most favorable for the chain reaction. In determining the nuclear safety of a chemical and metallurgical processing plant, all aspects of reactor theory may be involved, including both moderated and unmoderated systems; the problems are further enhanced because of the lack of certain experimental data and the uncertainty which exists in the theoretic values.

This paper is concerned primarily with nuclear safety considerations in the handling and processing of plutonium. Several questions are these: What types of criticality problems are encountered in a typical plutonium processing and metal fabrication plant? What criticality data are needed, and what are some of the methods and procedures which are used to achieve nuclear safety in operation?

### NUCLEAR SAFETY CONSIDERATIONS

Consider the conditions under which a given array of plutonium and other materials will become chain reacting. The exact configuration or spatial density must be known for each kind of atom present in the system. Thus, criticality depends not only on the amount of fissile material present, but on the size, shape, and material of any containment vessel which may be used, on the nature of any solvents or diluents, and on the presence of materials which may act as neutron reflectors.

In evaluating the nuclear safety of a given process, consideration must be given to the various in-plant factors which affect the criticality of an individual unit or quantity of fissile material, and due allowances made for the effects of neutron reflectors. The effects of neutron interaction between associated vessels or units in related operations (storage arrays, interconnected pipes, operations in adjoining hoods, etc.) must also be considered.

Some of the methods which have been used for criticality control are listed below; in some cases combinations of several different methods are used together in order to obtain the most efficient operation consistent with nuclear safety.

1. Process vessels are designed to be safe by geometry, in which case the neutron leakage is so high as to prevent criticality regardless of the quantity of fissile material contained therein.
2. Operating limits are imposed on material compositions, such as the density of fissile material contained per unit of a solution volume, or on the quantity of diluent or moderating material which may be present.
3. The amount of material handled in a single operation is limited to a subcritical amount, in which case a safe mass limit, or method of batch control is applied.
4. Restrictions are placed on the amount and form of material permitted in a given storage array, and on the distribution in space of the material contained within the array.
5. Soluble, or fixed poisons may be used in some cases to increase the mass limit, or to make a vessel safe by geometry.

## TYPICAL FLOW DIAGRAM - PLUTONIUM SOLUTION AND PLUTONIUM METALLURGY FACILITY

A typical flow diagram (Figure 1) has been prepared to illustrate the types of criticality problems which are encountered in handling plutonium and to indicate the areas in which they occur together with the principal methods of criticality control.<sup>(1)</sup> The preparation of  $\text{PuO}_2$  and any Pu metal from a  $\text{Pu}(\text{NO}_3)_4$  solution is not part of the plutonium fabrication plant operations.<sup>(2, 3)</sup> As far as these operations are concerned, the criticality problems begin with the receipt of the  $\text{PuO}_2$  or the Pu metal. The diagram is not intended to be a detailed, all inclusive, flow pattern of each step in the fabrication of the fuel elements.

In examining Figure 1, it is apparent that the criticality problems involve both homogeneous and heterogeneous systems, and include both moderated and unmoderated (fast systems), as well as lattices or bundles of fuel rods in a moderator (autoclave and etching process).

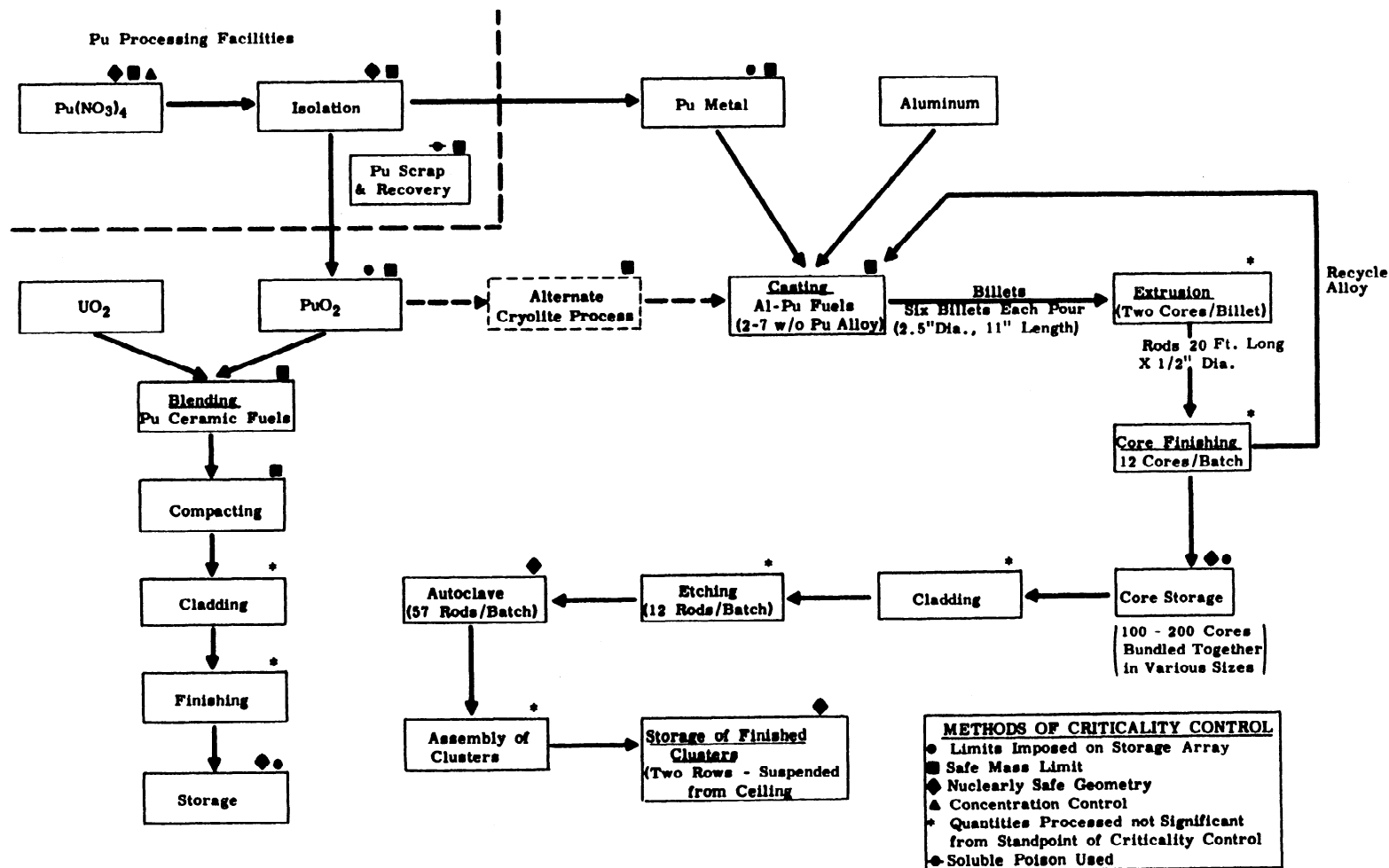
Further details will be given on the methods used for criticality control in a later section of this paper.

## CRITICALITY DATA AS A BASIS FOR NUCLEAR SAFETY

Data on plutonium criticality, which are used as a basis for establishing nuclear safety, are included and summarized in LAMS-2415.<sup>(4)</sup> Recommended nuclear safety limits are given in TID-7016 REV1.<sup>(5)</sup>

As noted in the references to these documents, criticality data on plutonium systems have been obtained primarily from the results of experiments conducted at the following U. S. sites: Data on plutonium metal were obtained from the Los Alamos Scientific Laboratory, the Lawrence Radiation Laboratory, and some data from the Rocky Flats Plant. Data on plutonium solutions were obtained from experiments conducted at Hanford.<sup>(6)</sup>

The nuclear safety of Pu solutions has been based on the results of the Hanford experiments, under which the criticality of aqueous Pu solutions was studied for plutonium concentrations up to 136 g/l.



**FIGURE 1**  
 Typical Flow Diagram - Plutonium Metallurgy Facility  
 Fabrication of Aluminum - Plutonium Fuels and Plutonium Ceramic Fuels

Some of these experimental data are presented in Figure 2, which gives the relationship between critical mass and volume for water tamped spheres and cylinders of aqueous  $\text{Pu}(\text{NO}_3)_4$  solutions. For the data shown, the Pu contained about three percent  $\text{Pu}^{240}$ , and the nitrate concentration was in the range of about 100-170 g/l depending on the concentration of the Pu. The  $\text{Pu}^{240}$  content of the bare sphere was about four percent. The critical mass values shown are for the total plutonium, including the  $\text{Pu}^{240}$ .

The effect of nitrate on the critical mass of water tamped spheres is shown in Figure 3.

The Hanford experiments also included measurements to determine the effect of  $\text{Pu}^{240}$  on the criticality of dilute Pu solutions in the range of 0.54-4.40 percent  $\text{Pu}^{240}$ .

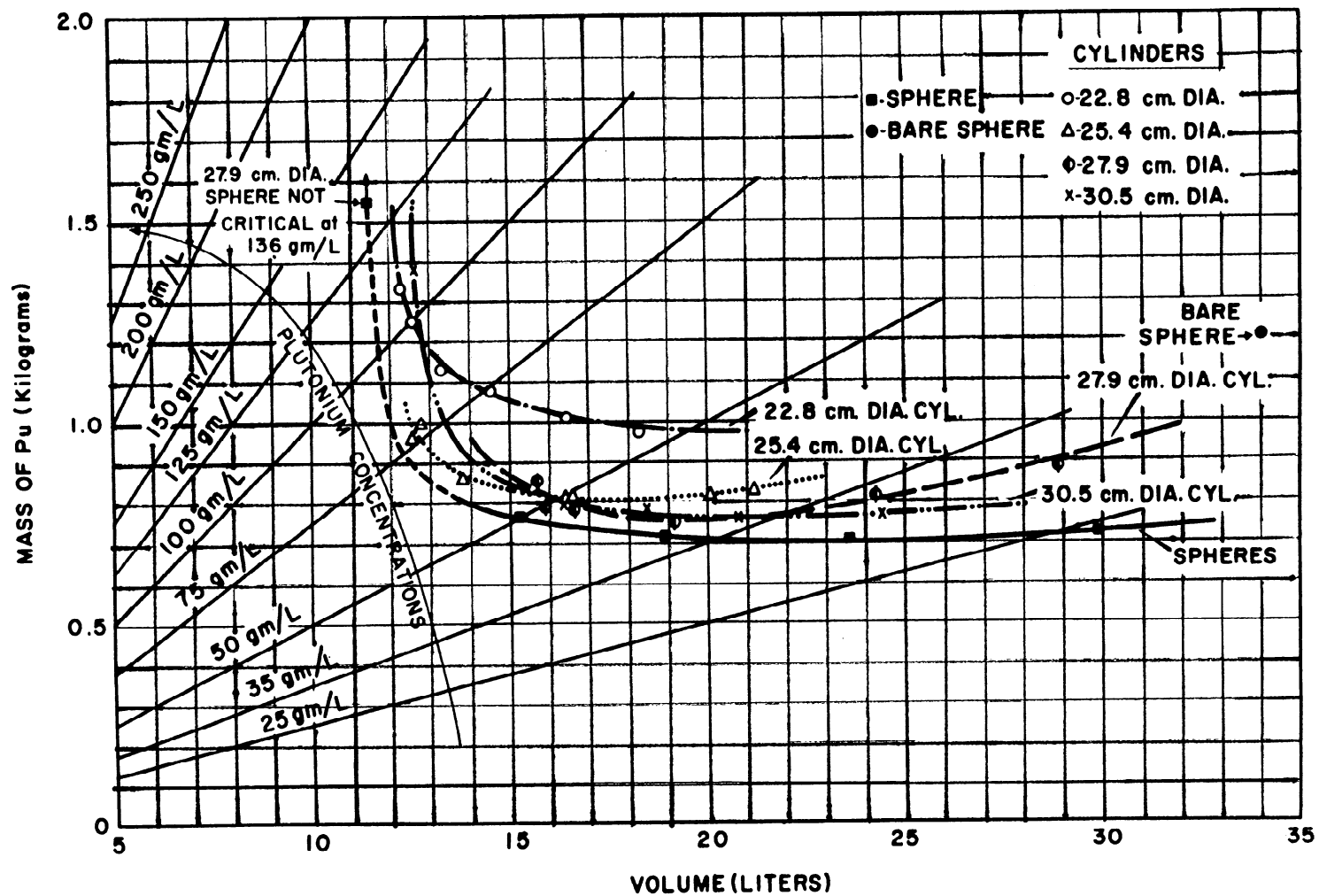
From the Hanford data, the minimum critical mass for a homogeneous Pu-water system was estimated to be 509 grams of  $\text{Pu}^{239}$ . The minimum value was obtained for a concentration of 33 grams Pu/l in a water reflected sphere about 12 inches in diameter.

The results of subcritical multiplication measurements have recently been reported in which Pu metal and Plexiglas plates were stacked together in cylindrical geometry.<sup>(7)</sup> These data may be used to provide a qualitative estimate of the critical mass in the intermediate range of moderation between solutions and solid metal.

For unmoderated  $\alpha$  phase Pu metal, the minimum water reflected critical mass is given as 5.6 kg.<sup>(5)</sup>

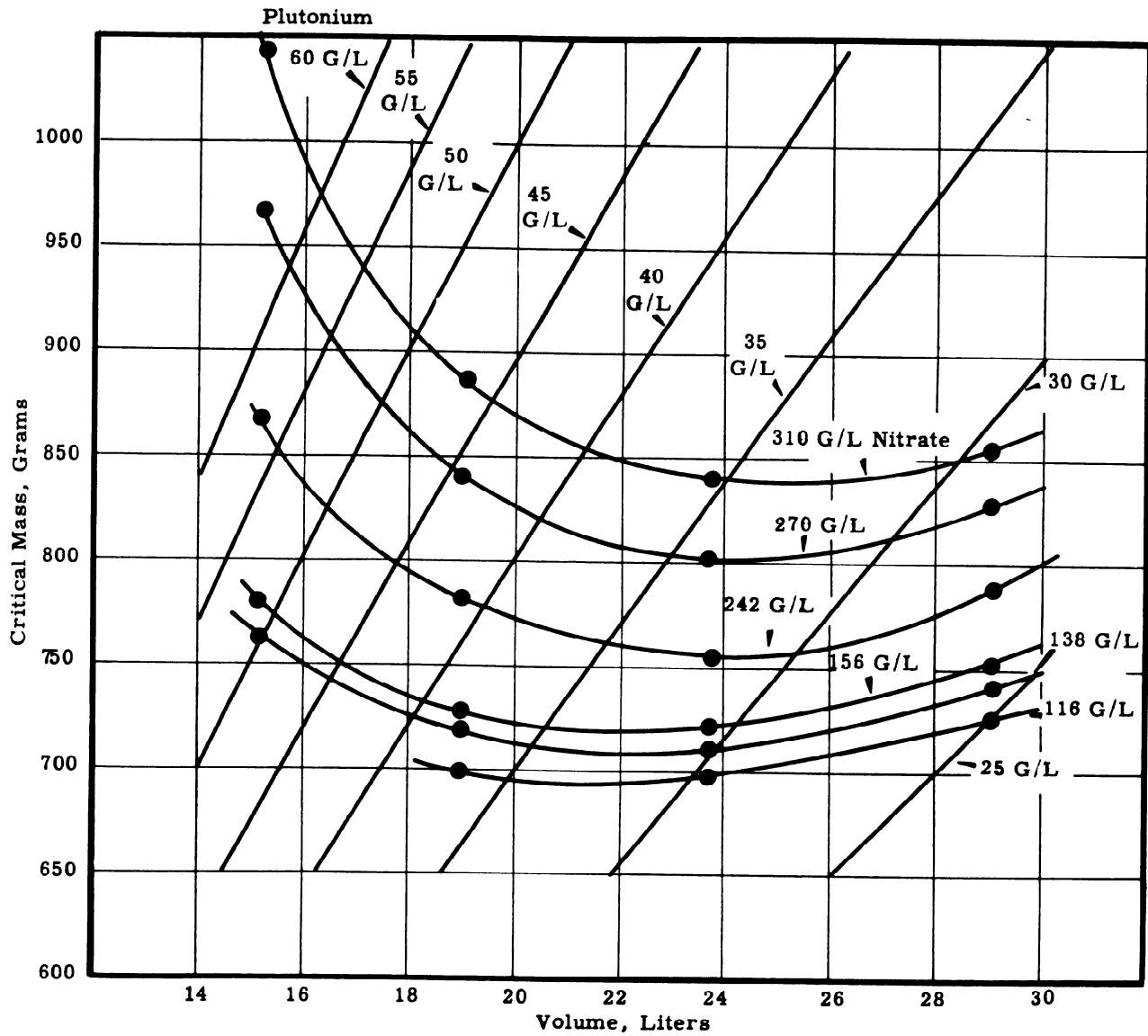
#### Criticality of Homogeneous $\text{PuO}_2$ -Water System

Criticality parameters for idealized Pu-water mixtures provide the bases for establishing nuclear safety in handling and processing plutonium, however, it is the homogeneous  $\text{PuO}_2$ , and  $\text{Pu}(\text{NO}_3)_4$  systems which are more frequently encountered.



**FIGURE 2**  
Critical Mass and Volume of Reflected Spheres and Cylinders  
of  $\text{Pu}(\text{NO}_3)_4$  in Aqueous Solutions





**FIGURE 3**  
Critical Mass versus Volume of  
Water-Tamped Stainless Steel Spheres,  
3.12 Percent Pu<sup>240</sup>, 27 C

Compounds, such as organic complexes, plutonium polymer, and  $\text{PuO}_2$ , may be formed nonroutinely in the aqueous parts of a plutonium nitrate process stream as the result of abnormal operations. For example, plutonium oxide may form if a nitrate solution contacts the wall of a high temperature evaporator, or the polymer may form if a nitrate solution at the proper pH is diluted or heated. (8, 9)

Information on the density and composition of plutonium polymer is meager, however, the density evidently depends greatly on the conditions under which the polymer is formed (i.e., pH, temperature, concentration, etc.). It has been postulated that a density of 1-2 g/cc may be possible; however, until there is further laboratory data, the maximum density that might be encountered can only be assumed. The formula for the precipitated polymer has not been firmly established. Experimental work at Hanford indicates that it is of the type  $\text{PuO}_2 \cdot X \text{H}_2\text{O}$  and could probably be,  $\text{PuO}_2(\text{H}_2\text{O})_{7 \frac{1}{2}}$ .

These "nonroutine" compounds have been found in certain parts of the processes in the past, however, there has been insufficient analytical data for positive identification. Identity has therefore been based on the nature of the process and the abnormal conditions encountered.

Plutonium dioxide is a product routinely formed as part of the metal isolation process; the oxide may also be used as a feed material for plutonium metallurgy in fuel element fabrication.

Some Pu density- $\text{H}_2\text{O}$  moderation relationships have been determined by mixing  $\text{PuO}_2$  with water. (10) These relationships are shown in Figure 4, together with the H/Pu ratios for Pu-water mixtures. The density of the  $\text{PuO}_2$  water mixtures is considerably less than that of the Pu-water mixtures at the smaller H/Pu ratios (for an H/Pu ratio of 4, the density of Pu in the oxide water mixture is  $\sim 3.5$  as compared with  $\sim 5$  for the Pu-water system).

A series of criticality calculations have recently been made for Pu systems using a one-dimensional, multigroup neutron diffusion code. (11) Eighteen energy groups were used in the calculations, which were performed

by Dr. W. A. Reardon with the use of the IBM-7090 digital computer.<sup>(12)</sup> The results of some of these calculations are presented in this and the following sections.

The calculated critical mass for a  $\text{PuO}_2$ -water system is shown in Figure 5; the density of the  $\text{PuO}_2$  for  $\text{H}/\text{Pu} = 0$  was assumed to have a theoretic value of 11.46 g/cc. The critical masses for the measured densities presented in Figure 4 are shown in Figure 6, and in Figure 7, the calculated critical diameters of infinite length cylinders are given for these densities.

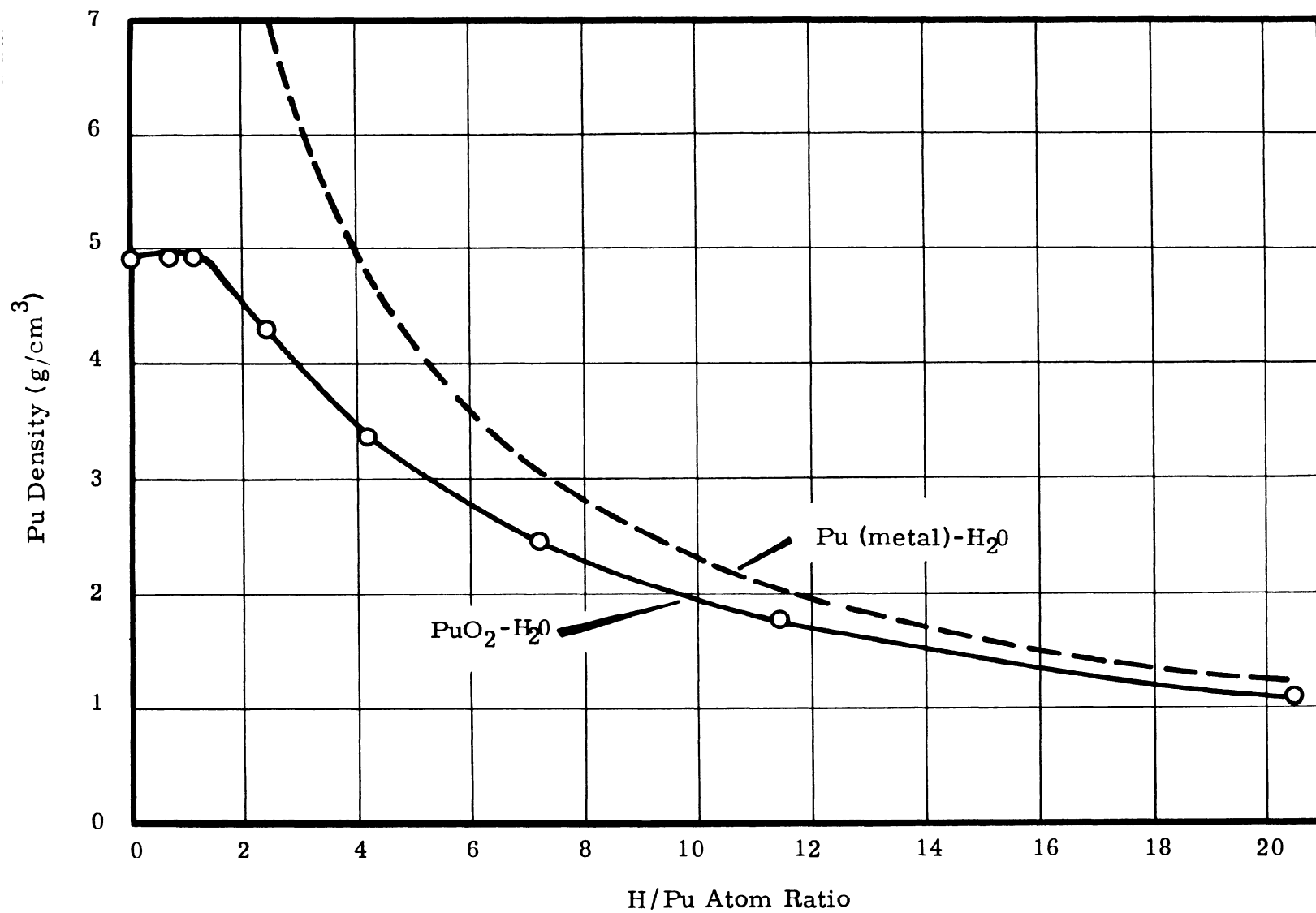
The large differences in critical mass between that of a Pu and  $\text{PuO}_2$  system is the result of the smaller density for the oxide system at the smaller  $\text{H}/\text{Pu}$  ratios.<sup>(4)</sup> The effect of density is also quite evident from a comparison of the calculated critical mass for  $\text{PuO}_2$  with a theoretic density of 11.46 with that for the  $\text{PuO}_2$  with a density of 5.6 at an  $\text{H}/\text{Pu}$  ratio of zero.

It is to be emphasized that the  $\text{PuO}_2$  results have not been confirmed by experiments. The calculations were made to indicate the large increase in critical mass which results because of the reduced density of the  $\text{PuO}_2$  and to provide improved estimates of the criticality parameters of these systems.

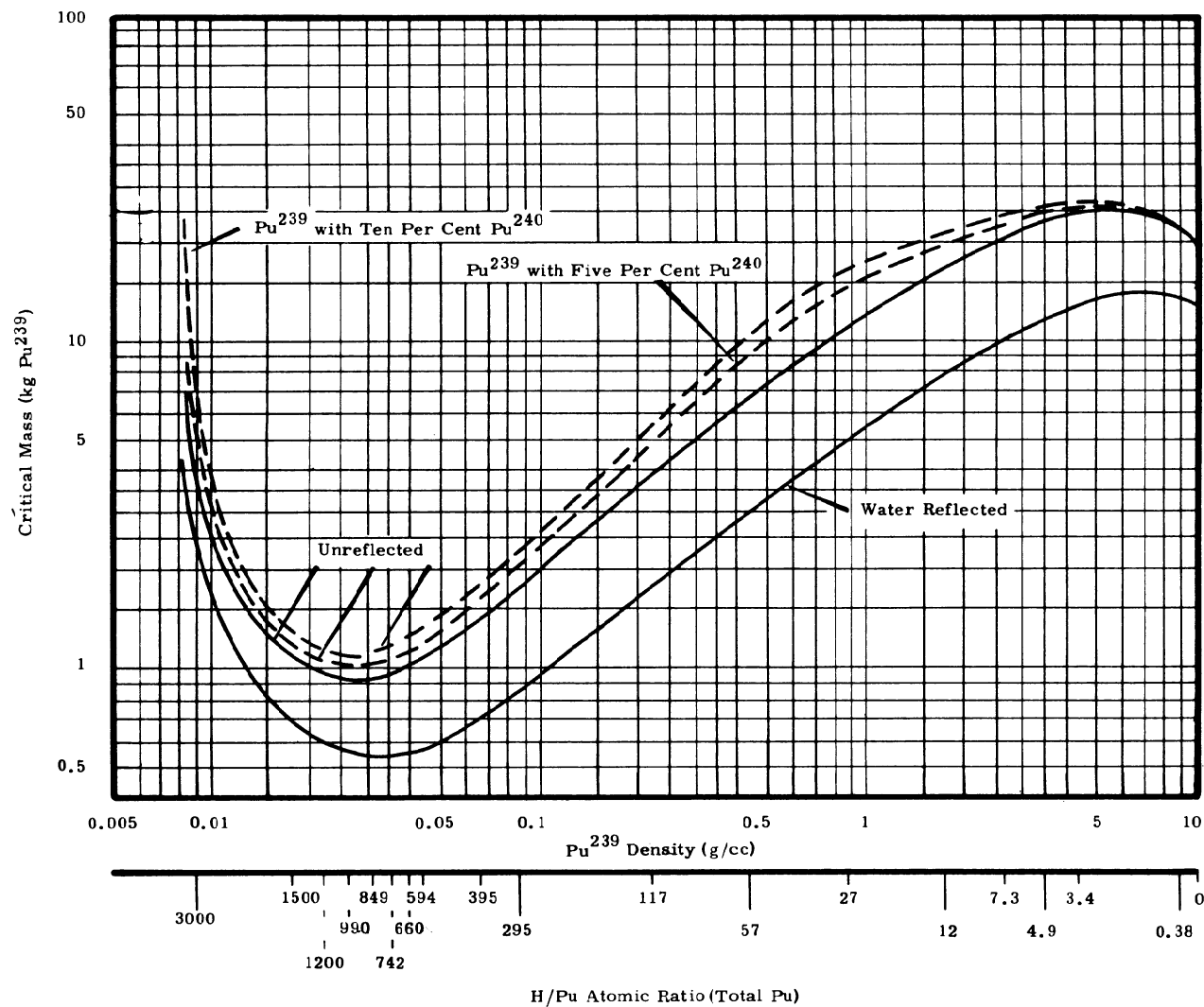
#### Effect of $\text{Pu}^{240}$ on Criticality

The Hanford P-11 experiments provided data for the effect of  $\text{Pu}^{240}$  on the critical mass of dilute Pu solutions for isotopic content ranging up to about four percent  $\text{Pu}^{240}$ .

These experimental results have been calculated with the 18 group cross section set devised for the 9-Zoom code. Excellent agreement was obtained between the measured and calculated effect of  $\text{Pu}^{240}$  on the critical mass for the 14-inch water tamped sphere with  $\text{H}/\text{Pu}$  atom ratios in the range of 824-953.<sup>(6)</sup>

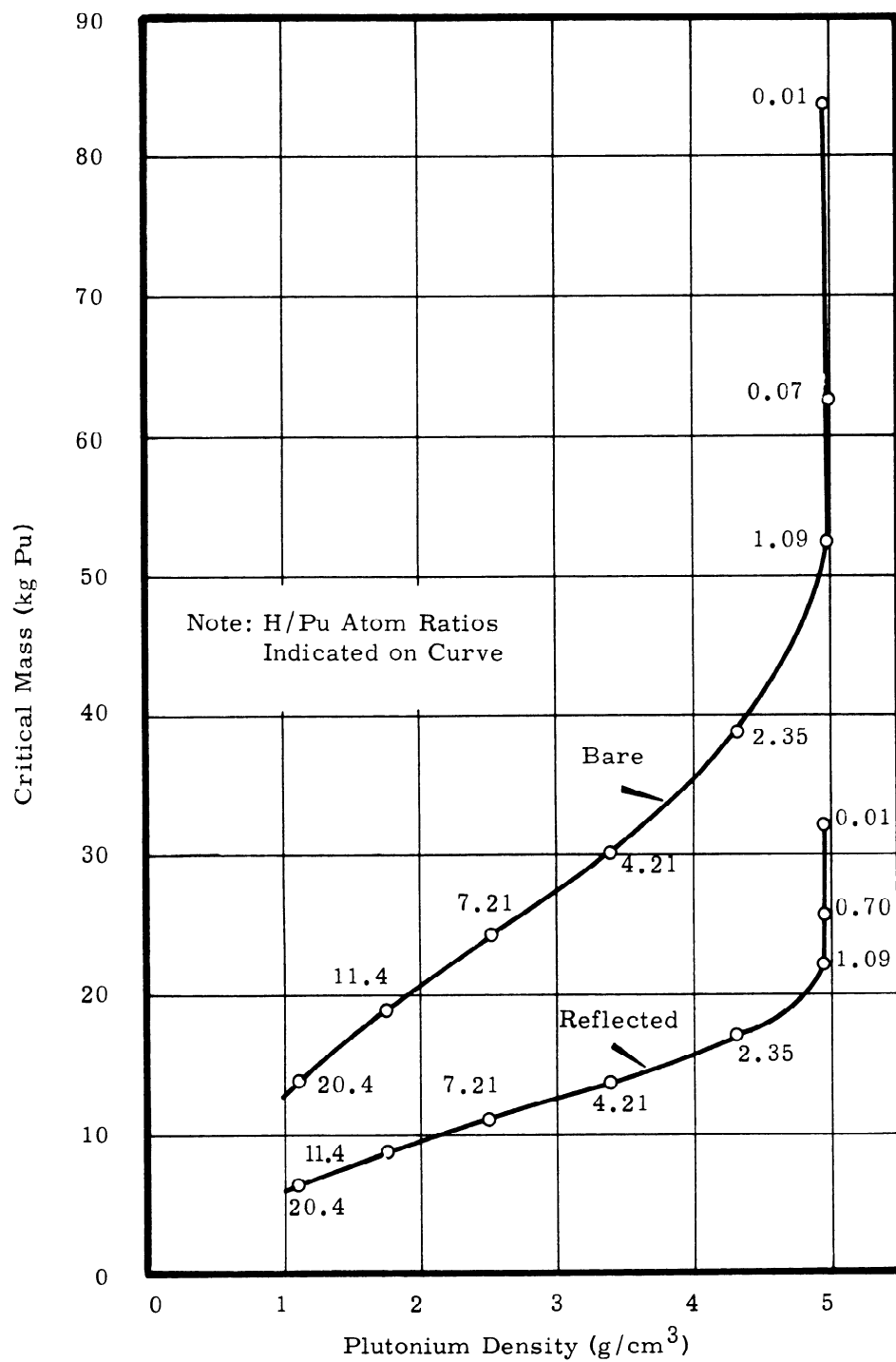


**FIGURE 4**  
Plutonium Concentration versus Hydrogen-to-Plutonium Atom Ratio



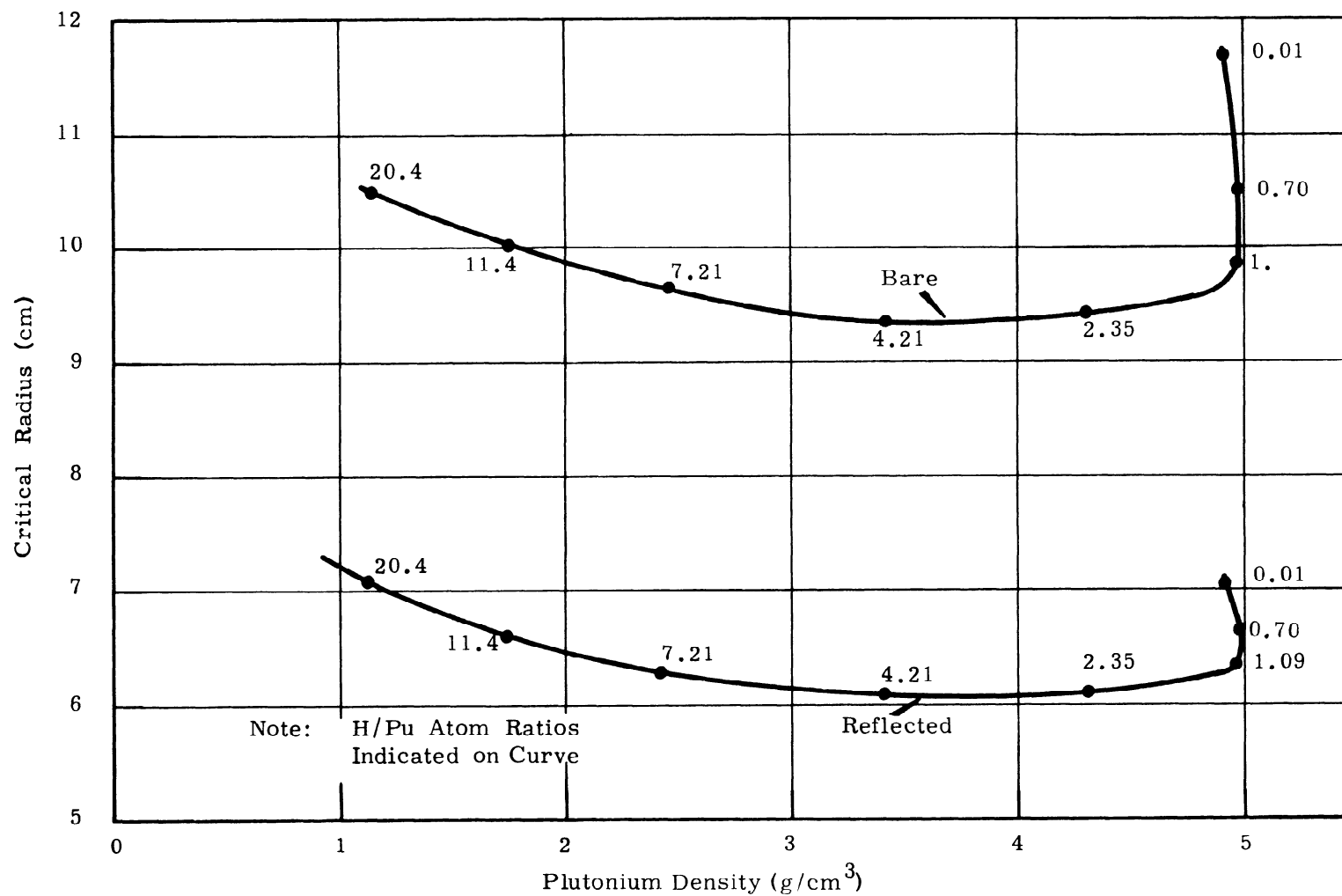
**FIGURE 5**

Estimated Critical Mass of Water Moderated Plutonium Oxide Spheres



**FIGURE 6**

Estimated Critical Mass of Water Moderated Plutonium Oxide Spheres  
(Based on Measured Density of PuO<sub>2</sub>)



**FIGURE 7**  
 Estimated Critical Radius of Water Moderated Plutonium Oxide Infinite Cylinders  
 (Based on Measured Density of PuO<sub>2</sub>)

The results of these calculations are summarized briefly in the following table:<sup>(12)</sup>

<u>Calculated <math>k_{\text{eff}}</math> for 14-inch Water-Tamped Sphere of <math>\text{Pu}(\text{NO}_3)_4</math> (<math>\sim 110</math> g Nitrate/l)</u>			
<u>Percent <math>\text{Pu}^{240}</math></u>	<u>H/Pu Atom Ratio</u>	<u>Critical Mass from Measurements<sup>(6)</sup></u>	<u>Calculated <math>k_{\text{eff}}</math> of Critical Assembly</u>
0.54	824	626 g $\text{Pu}^*$	1.0000
1.76	860	656	1.0010
3.12	911	700	0.9942
4.05	953	720	0.9987

\* Including  $\text{Pu}^{240}$

To be in agreement with experiment, the calculated  $k_{\text{eff}}$  should be unity in each case.

The effect of  $\text{Pu}^{240}$  has been calculated for water-moderated  $\text{PuO}_2$  spheres with five percent and ten percent  $\text{Pu}^{240}$ ; the results are shown in Figure 5.

The calculations for higher concentrations, for example H/Pu ratios less than 50, are extremely sensitive to the effective resonance absorption cross section for the  $\text{Pu}^{240}$ , and are therefore not expected to be reliable. The  $\text{Pu}^{240}$  acts as a neutron poison; the calculations indicate the  $\text{Pu}^{240}$  to have the greatest effect on the critical mass for H/Pu ratios in the range of 20 to 60, decreasing at the smaller H/Pu ratios. In the case of the Hanford experiments with the 14-inch sphere, the critical concentration of  $\text{Pu}^{239}$  was increased by about three percent for each percent  $\text{Pu}^{240}$  present.<sup>(6)</sup>

Measurements made at Los Alamos in fast critical assemblies indicate that the net fission production of  $\text{Pu}^{240}$  is about 2/3 that of  $\text{Pu}^{239}$ . It is also reported that the  $\text{Pu}^{240}$  present in  $\text{Pu}^{239}$  metal to less than  $\sim 10$  percent increases the critical mass slightly.<sup>(13)</sup>

Experimental data on the effect of  $\text{Pu}^{240}$  will be determined as part of the experimental program for the new Hanford Plutonium Critical Mass Laboratory (a description of the proposed program is included in a later section of this paper).<sup>(14)</sup>



### Nuclearly Safe Concentration for Aqueous Plutonium Solution

A quantity of special interest relative to the nuclear safety of solutions is the nuclearly safe concentration limit, i.e., the maximum permissible concentration of fissile material for which  $k_{\infty} \leq \text{unity}$ . \*

Recent measurements have been made of  $k_{\infty}$  for dilute plutonium nitrate solutions in the Hanford Physical Constants Testing Reactor. Some additional confirming experiments are required before the results will be finalized. However, the current data indicate, after corrections are applied for the effects of  $\text{Pu}^{240}$  and nitrate, the limiting concentration to be about  $9.1 \pm 1$  g Pu/l for a homogeneous  $\text{Pu}^{239}$  water system. This number is about 1-2 g/l higher than the calculated value. The reasons for the disagreement have not been determined.

### Nuclearly Safe Ratio of Plutonium-to-Uranium

If  $\text{U}^{235}$  is diluted sufficiently with  $\text{U}^{238}$ , there exists an atom ratio beyond which criticality can not occur because of the excess neutron absorption in the  $\text{U}^{238}$ . The limiting enrichment of uranium below which criticality is not possible in an aqueous homogeneous system is 1.0 percent, i.e., when the  $\text{U}^{235}$  is one weight per cent (w/o) of the total uranium. <sup>(15)</sup> There is also a plutonium-to-uranium (Pu/U) ratio below which criticality is not possible for a homogeneous system. The ratio for plutonium has not been measured as in the case of uranium, but has been calculated as, 0.0018. This Pu/U atom ratio is believed to be sufficiently conservative for use as a basis in criticality control; the number implies that adding one atom of Pu to natural uranium would be the equivalent of adding about 1.7 atoms of  $\text{U}^{235}$ . The ratio should be recalculated in view of the improved cross sections which have become available since the original calculation was made. As mentioned in a later section, the ratio of 0.0018 has been used as a basis for critical control under certain conditions, but with a safety factor applied.

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\* Sometimes referred to as the "infinite sea" concentration. For concentrations below this value criticality will not occur because of excess neutron absorption in the hydrogen.

# Heterogeneous Systems of Pu-Al Fuel Rods in Light Water<sup>(16)</sup>

A series of critical approach and exponential experiments were conducted at Hanford with Pu-Al alloy fuel elements in light water moderated lattices. The fuel elements were Al - 5 w/o Pu, 0.506-inch in diameter and 24 inches in length.

The work was done in connection with the Plutonium Recycle Program to provide critical mass data relevant to the processing and handling of this kind of fissile material. The data are of use in connection with nuclear safety specifications in the fabrication of fuel elements and for correlating critical mass calculations with experiments for heterogeneous Pu systems.

The fuel rods were clad in 0.030-inch Zircaloy-2 with a 0.020-inch and 0.125-inch thick end caps. This gave Zr/Pu atomic ratio of 31.92 and an Al/Pu atomic ratio of 168.20. A hexagonal pattern was used for the lattices which were fully water reflected.

The minimum critical mass for the five percent plutonium enriched Pu-Al alloy rods in spherical geometry was about 1.5 kg of Pu (including Pu<sup>240</sup> and higher isotopes) for an H/Pu ratio of ~ 650. The maximum buckling for these systems was about  $11,300 \times 10^{-6} \text{ cm}^{-2}$  for an H/Pu ratio of ~ 450.

Data were obtained on the extrapolation lengths for the reflected assemblies by equating the buckling expression for the exponential measurements with the critical buckling as determined from the critical approach measurements.

When the results of the measurements are corrected for the effect of Pu<sup>240</sup>, the minimum critical mass is estimated to be about 1.4 kg Pu<sup>239</sup>.

These data have been used as a basis for extending the theory to calculate mass limits of isolated units of Pu-Al alloy rods in water for several other enrichments and diameters; some of these results are given in the following table:<sup>(17, 18)</sup>

Safe Mass Limits for Pu-Al Alloy Rods

<u>Enrichment (Weight Percent Pu in Alloy)</u>	<u>Rod Diameter (Inch)</u>	<u>Safe Mass Limit* (kg Pu)</u>
5.0	0.25	0.39
	0.50	0.52
	0.75	0.65
15.0	0.25	0.35
	0.50	0.61
	0.75	1.00

\* The safety factor is about 2.3 for double batch allowance.

Plutonium-Aluminum Alloy - Unmoderated System

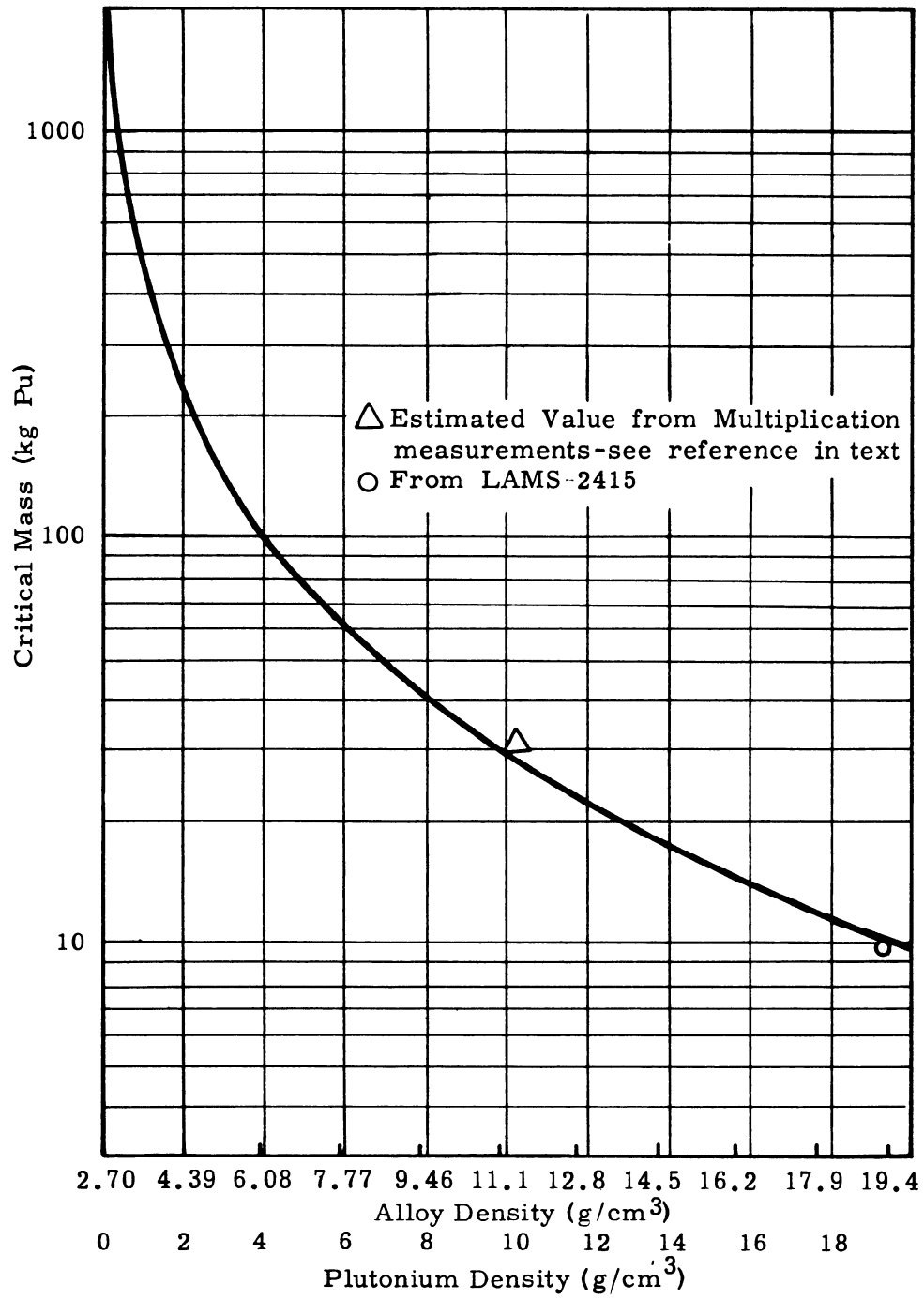
Allowance factors on mass limits are given by Woltz, et al.<sup>(5)</sup> which may be used to determine a "safe" mass for plutonium when mixed homogeneously with other elements (excluding H, D, Be).

The mass limit for Pu-Al alloy is of special interest in connection with the fabrication of Pu-Al fuel elements for the Plutonium Recycle Program.

To provide better estimates than currently available, the eighteen-group diffusion code (9-Zoom) was used to calculate the critical mass for Pu-Al alloys.<sup>(12)</sup> The high energy transport cross sections of plutonium were adjusted to provide approximate agreement with the critical mass of  $\alpha$  phase Pu at full density. The results are shown in Figure 8. The calculations are in qualitative agreement with the estimated spherical critical mass for a nominal volume ratio of plutonium to aluminum of 2:1 (Pu density  $\sim 9.7$  g Pu/cm<sup>3</sup>).<sup>(19)</sup>

Plutonium Metal Dissolution

The dissolution of plutonium metal involves a system of one or more pieces of plutonium, which during the course of dissolving are immersed in Pu solution. Such a system is encountered in metal scrap recovery operations.



**FIGURE 8**

Estimated Critical Mass of Plutonium-Aluminum Alloy Spheres  
(Unreflected)

For the case of plutonium metal pieces in a moderator, it is not possible to give general recommendations concerning nuclear safety, but there are some applicable data which may be used to arrive at answers.<sup>(4)</sup>

A study has been made also, which indicates that a plutonium solution-plutonium metal system might be more reactive than a plutonium metal-water system. The analysis was based on highly enriched uranium metal immersed in uranium solutions. The analysis indicates that a piece of fissile material, which is subcritical when placed in a solution of nitric in which the solution height is safe for the completely dissolved metal, might become critical during the process of dissolution when the metal is partly dissolved and partly in solution.<sup>(20)</sup> Additional experiments are required to check these conclusions.

#### APPLIED METHODS OF CRITICALITY CONTROL IN PLUTONIUM PROCESSING AND FUEL ELEMENT FABRICATION

Some of the methods which have been used for criticality control in plutonium processing and metallurgy operations are discussed below.

##### Plutonium Solutions

##### Nuclearly Safe Geometry

The preferred method of criticality control is to design vessels which are safe by virtue of their geometry. Because of the excess neutron leakage, vessels so designed will remain subcritical regardless of the amount of fissile material contained therein. Thus, no further criticality control is required providing the integrity of the vessels is maintained. In determining the always-safe dimensions, allowances must be made for the effects of neutron reflectors which may be proximate, and for possible neutron interaction effects from adjacent vessels, which can cause a reduction in the geometrically safe dimensions of an isolated unit.

### Safe Batch or Mass Limit

The amount of plutonium that is placed in a container which is not geometrically safe is limited to a safe batch, or mass limit. This amount of plutonium which is less than the minimum critical mass, is determined from analysis, and volume or weight measurements.

Some examples of batch limits are the following:

1. The amount of Pu is limited to less than 1/2 (45 percent) of the minimum critical mass. (Safe for double batch allowance.)
2. The amount of plutonium contained in the solution is limited to a fraction of the minimum critical mass for the vessel assuming a uniform water-tamped precipitate layer. (The vessel is then also safe should precipitation occur.)

### Limiting Critical Concentration of Plutonium

The limiting critical concentration of plutonium which has been used as a basis for criticality control in aqueous solutions is 7.8 g Pu/l.<sup>(5)</sup> For concentrations less than 7.8 g/l ( $H/Pu \geq 3600$ ), excess neutron absorption in the hydrogen prevents criticality even for an infinite system.

The maximum plutonium concentration which has been used at Hanford in large vessels for purposes of criticality control is 6 g Pu/l (23 g/gallon).

The degree of safety in using the value of 6 g Pu/l is larger than implied, since in any finite vessel the critical concentration will be greater than 7.8 g/l; also the effect of  $Pu^{240}$ , nitrate, etc., would result in a further increase in the minimum critical concentration.

The maintenance of a subcritical concentration is not in itself considered adequate safety against criticality in large vessels. Consideration must also be given to the possibility that plutonium might be precipitated from solution because of operational errors. Limits are, therefore, also established for the total amount of plutonium contained in vessels which are not safe by geometry.

Whenever there is a possibility that plutonium precipitation might occur, a mass limit is established for the vessel. This limit is a fraction of the calculated minimum critical mass for the vessel on the basis of a uniform precipitate layer.

In the case of solutions, control is effected through adjustment of the stream ratio to obtain plutonium concentrations corresponding to the storage of no more than an arbitrarily chosen amount (90 percent) of the calculated minimum critical mass when the vessel is filled to the overflow, or the solution transfers are limited so that vessels never contain more than that fraction of the calculated minimum critical mass.

#### Plutonium Solutions with High Uranium Concentrations

The always-safe ratio of plutonium-to-uranium has been used as a basis for criticality control of solutions which are high in uranium concentrations. Stream composition specifications have been based on a maximum ratio of 0.0017 (1500 g Pu/ton of U) for vessels in which this method of criticality control is applied. The calculated critical ratio for an infinite system is, as given previously, 0.0018. There are additional margins of safety in using the value of 0.0017, such as the effect of  $\text{Pu}^{240}$ , and other neutron absorbing materials (i.e., nitrate) which might be present in the solution. Also, the maximum value of  $k_{\infty}$  does not occur for H/Pu ratios characteristic of solutions, but for those ratios more nearly typical of slurries and precipitates. Thus, in the absence of precipitation, the H/Pu ratio of 0.0017 includes a large safety factor for normal conditions with homogeneous solutions. Note that in applying the above ratio it is assumed that if precipitation were to occur, the ratio would not be changed. Experiments have shown that under the proper conditions, the plutonium concentration relative to uranium in a precipitate may be increased by as much as a factor of two.<sup>(21)</sup> Therefore, steps should be taken to insure that conditions favoring precipitation do not occur, such as a system with low acidity, etc. (In case of plutonium nitrate solutions there will be no problem if the plutonium has first been oxidized to the hexavalent state, for in this case the ratio will not be increased even if precipitation does occur.)

### Soluble Poisons

In those cases in which a soluble poison (cadmium) has been used at Hanford, its use has been primarily that of a secondary control to safe plutonium solutions, but not as a primary control. When the poison is used in this manner, criticality will not occur if the poison is removed from the process unless some other operating error were to occur simultaneously, such as double batching or exceeding some preset concentration limit.

### Plutonium Fabrication Plant

#### Plutonium Storage

The number of units permitted in a storage array depends on the size of each unit in the array and on the separation between units.

Criticality data which may be used as a guide for determining the nuclear safety of arrays are given by Paxton,<sup>(4)</sup> and criteria for storage is presented by Woltz, et al.<sup>(5)</sup>

Figure 9 is an example of a safe storage system used for Pu metal and unmoderated  $\text{PuO}_2$ ; the center-to-center distance between units is  $\geq 24$  inches. This is an interesting method of storage, since two units in adjacent compartments can not be placed together. The center-to-center spacing between units in different compartments (24 inches in this case) is always assured.

The maximum unit of Pu per compartment is clearly posted, and detailed records are maintained of all additions and removals from the array.

### Casting Operation - Plutonium-Aluminum Alloys

One of the processes used in making Pu-Al alloy is the direct addition of plutonium metal to molten aluminum. With no moderator present, the maximum amount of plutonium which may be alloyed with aluminum in a single batch is 2600 grams. This batch size is required during the alloying process since 100 percent plutonium is being alloyed with the aluminum. Safety can not be based on the alloyed material until after the alloying process is completed.



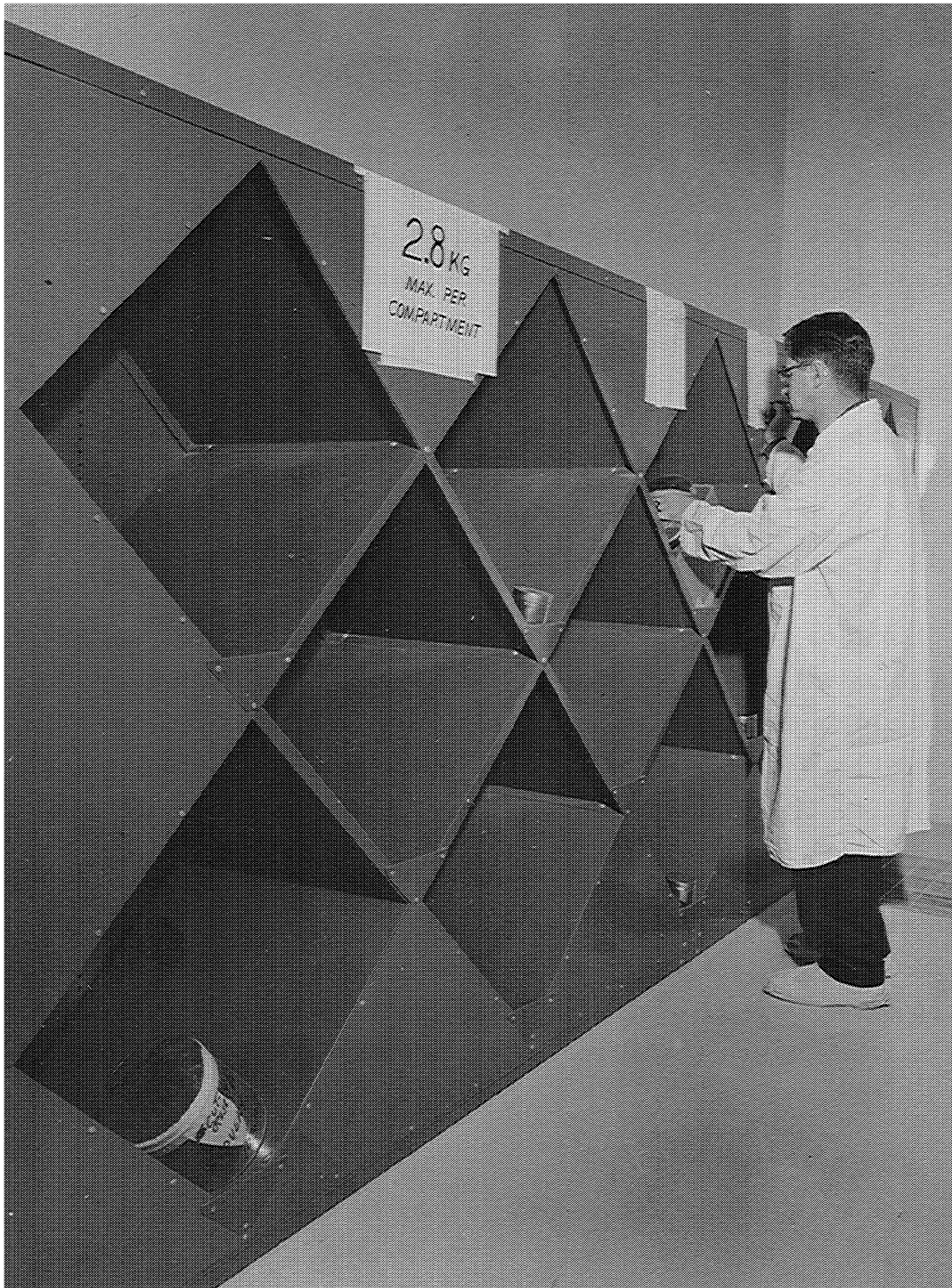


FIGURE 9  
Safe Storage Array of Fissile Material

### Cryolite Process

The cryolite process is also used in making the alloy. In this process,  $\text{PuO}_2$  is added to the molten aluminum along with cryolite flux. The aluminum reduces the  $\text{PuO}_2$  to plutonium metal and the alloying takes place. Larger batch limits would be allowed in this operation, because the  $\text{PuO}_2$  has a significantly higher critical mass than the metal.

### Core Storage

The cores are stored in wooden boxes. Geometry is given primary consideration in establishing a safe limit for each box and on the number of boxes which may be safely stacked together.

### Autoclave

Nuclear safety of the autoclave is based on geometry control. Figure 10 shows a bundle of fuel elements being inserted in the autoclave. The diameter of the autoclave is less than the critical diameter for lattices of Pu-Al alloy rods in water for the plutonium enrichments being handled. In this case, the diameter is 6-1/2 inches (2-7 w/o Pu alloy).\*

### Storage of Completed Fuel Elements

The storage of completed fuel elements is based on geometry control. The fuel elements are stored in a safe slab arrangement, which would also be subcritical in the event of water flooding. Figure 11 shows such an arrangement. Two rows are permitted, although only one is shown in the figure.

### Hood Limits

Criticality control is effected by setting safe limits on each hood. These limits are usually conservative by more than a factor of two. Further control is afforded by the accountability procedures which require that all plutonium additions and subtractions be noted on the individual hood logs.

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\* Quite conservative; in view of recent experiments, the "safe diameter" may now be increased to nine inches with nearly double capacity.<sup>(16)</sup>

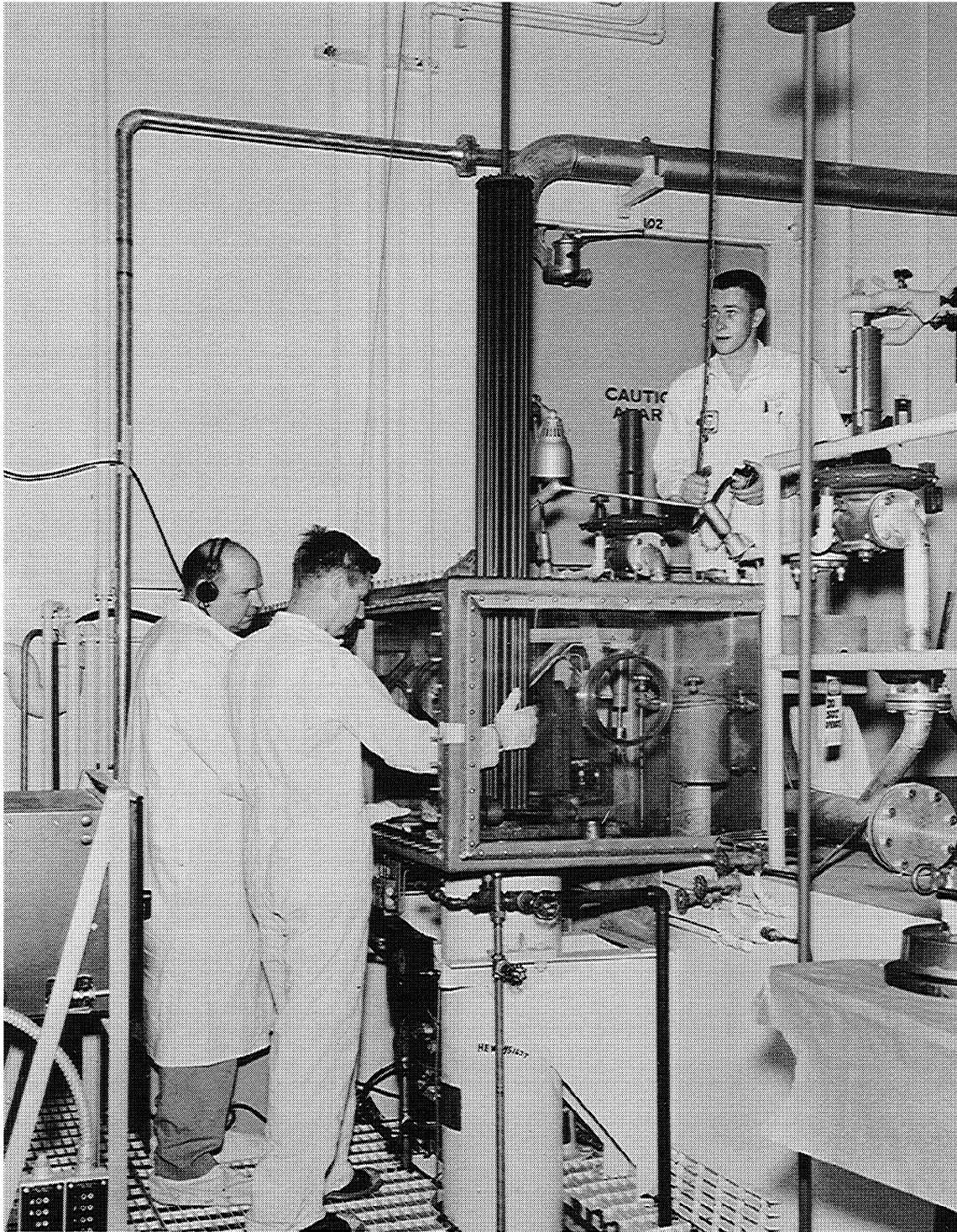


FIGURE 10

Fuel Tubes Being Inserted in Autoclave Which is Safe by Geometry



FIGURE 11

Method of Storing Fuel Rod Clusters in Safe Slab Geometry



Furthermore, the piping of water to fabrication hoods is avoided wherever possible to eliminate the possibility of accidental flooding, which could result in conditions being more favorable for a chain reaction to occur.

### SOME GENERAL TYPES OF CRITICALITY PROBLEMS FREQUENTLY ENCOUNTERED

A typical list of some general types of problems involving nuclear safety in chemical and metallurgical processing of plutonium is given below. These problems do not constitute a complete listing, but are representative only of those general types which usually require more than a preliminary examination for solution.

#### Some General Types of Criticality Problems

- Pairs or groups of cylinders
- Arrays of metal parts
- Arrays of various units (mainly powders and metal)
- Unusual reflectors or reflecting conditions
- Unusual shapes of solution vessels; unusual spacing and geometries with solid compounds
- Single units (usually solution cylinders); precipitates or slurries
- Unusual moderator, or nonhomogeneous systems, dissolution of metal
- "Safe" ratios or poisons
- Metal alloys
- Fuel element fabrication - storage and handling

### ADMINISTRATIVE PROCEDURES

A listing of some typical procedures which have been followed in effecting criticality control are given below:

1. Potential for critical mass incidents are examined at the time designs are being developed for new facilities, or for revisions to existing facilities, and this potential is minimized through appropriate design. Critically safe geometry is the preferred design criterion.

2. In situations where safety can not be assured by geometry, administrative procedures for control of the process operation within safe limits are established and enforced. Critical mass control under these procedures is based on operational controls such as dilution, neutron poisons, and batch size limits. These procedures are in the form of written specifications for the particular facility under consideration. Adherence to the written specifications is implemented by incorporation of specific instructions in operating procedure, by posting of critical mass control limits in appropriate locations, and through training of personnel.
3. When process equipment is altered or is replaced by equipment not identical with the original in design, the effects of the changes on critical mass control safety margins are re-evaluated. If the changes reduce the margins of safety provided by existing control procedures, these procedures are revised to re-establish the required margins of safety prior to introducing fissile materials into the equipment.
4. When batch size limits are used as a control method, these limits are such that inadvertent double batching will not result in a critical mass incident.
5. Transfers of unsafe concentrations of fissile materials from geometrically safe vessels to geometrically unsafe vessels are made only in carefully measured, subcritical batches or under controlled conditions of dilution to safe concentrations.
6. At each facility where there is potential for critical mass incident, there must be in existence a detection system, equipped with distinctive, audible alarms, which is capable of detecting promptly a radiation burst arising from a

critical mass incident. An evacuation procedure is prepared which is to be followed in case the alarm is sounded or if other warning of a critical incident is received. Personnel are trained to respond in accordance with these procedures.

7. Internal audits will be performed at the direction of the Operation Manager on a continuing basis to assure that administrative controls are adequate and adhered to.
8. A program for improving the analytical methods and measurements associated with critical mass control is maintained and improvements that are developed will be incorporated promptly into the standard analytical and measurements methods.
9. At each facility where critical mass control is required, training programs are maintained which keep personnel informed of the general principles on which critical mass control is based and of the operational precepts which must be followed to maintain control.
10. Each facility is required to have procedures for building re-entry following a critical mass incident.
11. When there is doubt that control is within established critical mass specifications, the facility is shut down until control can be assured.

#### PROPOSED GENERAL PROGRAM OF STUDIES FOR HANFORD PLUTONIUM CRITICAL MASS LABORATORY

A new critical mass laboratory has recently been completed at Hanford, Washington. This laboratory was built for the purposes of obtaining needed criticality data on plutonium solutions and precipitates of plutonium in support of chemical processing. The laboratory is fully described by Reardon, et al.<sup>(14)</sup>

The program for the laboratory is intended to be a general study involving homogeneous and heterogeneous experiments needed for obtaining criticality data on plutonium solutions and precipitates. A combined theoretical and experimental study will be made of "clean" cases, i.e., to those cases in which a decisive comparison between theory and experiment can be made. With this study as a guide, the more complicated cases, often encountered in plant design, will be investigated experimentally; the extent to which these types of experiments will be conducted depends on the degree of correlation which can be effected between theory and experiment for the simple geometries.

From a general viewpoint, the program will have a several-fold objective:

1. To supply critical mass data which will be in direct support of current processes and be of use in the design and operation of new radiochemical equipment.
2. To obtain a comprehensive set of critical mass physics data which will serve as the basic source of information for nuclear safety in the prevention or termination of inadvertent critical nuclear reactions in nonreactor environments.
3. To provide basic information on the criticality of plutonium systems and thereby assist in the development and testing of theory for plutonium reactor systems. As theoretical methods are developed and correlation effected between theory and experiment, an increasing number of plutonium criticality problems can be solved by theory - with significant reductions in the experimental program.

Initial geometries studied will be spheres and cylinders with eventual extension to slab geometry. A comparison of the critical dimensions for the different geometries will be useful in evaluating extrapolation lengths and



for determining if the relationships ("shape" factors) between plutonium systems in different simple geometries are consistent with  $U^{235}$  criticality data which have been obtained by the Oak Ridge National Laboratory group.

The general program of criticality studies includes the following list of experiments.

### Homogeneous Systems

#### 1. Experiments with Plutonium Solutions

Plutonium in some form of a water complex is the type of system most frequently encountered. Such systems were studied under the P-11 Program for plutonium concentrations up to 136 g/l.<sup>(6)</sup> In view of the current type of separations processes (continuous flow), data are now needed for higher concentrations of plutonium. As the plutonium concentration is increased, the neutron energy spectra becomes more characteristic of intermediate reactor system rather than of a thermal type. Critical mass studies are planned with plutonium solutions, and precipitates or homogeneous mixtures of plutonium with principal emphasis on H/Pu<sup>239</sup> ratios less than 400. The critical dimensions of spheres, cylinders, and slabs will be determined and "shape factors" evaluated for converting from spherical to cylindrical and/or rectangular geometry.

Neutron flux distributions will be measured in selected critical assemblies for evaluating bucklings and extrapolation lengths. Calculated thermal and epithermal flux distributions will be compared with measurements, and a study made of theoretic models for the critical systems in the thermal and intermediate energy range.

#### 2. Experiments with Precipitates of Pu or Facsimiles

Plutonium compounds in the density range 1-4 g/cc. and with H/Pu ratios of about 25-5 are encountered in certain operations of the separations plants. Some of these compounds are  $PuO_2$ ,

$\text{Pu}(\text{C}_2\text{O}_4)_2$  and  $\text{PuO}_2(\text{H}_2\text{O})_{7\frac{1}{2}}$ . The latter compound is the plutonium oxide polymer, which forms from plutonium nitrate solution under the proper conditions of acid molarity and temperature. In order to establish the criticality parameters for this plutonium density and H/Pu range, experiments will be made with  $\text{PuO}_2$ -plastic mixtures or other  $\text{PuO}_2$ -hydrogenous mixtures. The material will be in the form of solids, or compacts which have been sealed or canned for contamination control. A split table remote assembly machine will be used for these experiments.

### 3. Complex Geometry

While most of the experiments will be done with geometries for which a decisive comparison between theory and experiment can be made (spheres, cylinders, and slabs are examples of such forms), the safe mass limits will be determined for the "one-of-a-kind", odd-shaped vessels which exist in separations processes, for which only crude approximations can be made. These experiments will be done as required to evaluate the nuclear safety of the processes involved.

### 4. Interaction Experiments

Where more than a single subcritical unit of fissile material is handled, there is always the probability of interaction between units. It must be determined in all instances that there is no criticality hazard. Theoretical calculations do not give reliable answers in many such problems, and thus experiments must be performed.

It is expected that "interaction" experiments will be conducted for arrays of cylinders and for interacting parallel slabs. In these experiments, work will be done with solutions and precipitates of plutonium with bare and partially reflected units.

5. Neutron Reflectors

The effect of a given reflector on the critical mass of a system will differ depending on the median energy of neutrons in the system and on the position of the reflector relative to others which may be present. As an example, a thin stainless steel wall of a reactor vessel will reduce the effectiveness of a water reflector, whereas a thick layer of stainless steel may be equivalent of, or an even better reflector than that of water.<sup>(22)</sup> For such reasons, many of the critical mass determinations will be of reflected systems. Of interest are the effects on the criticality of various reflectors, such as concrete, water, polyethylene, stainless steel, etc., as well as experiments to determine the effective reflector savings for an "equivalent man".

6. Effect of Density on Criticality

Critical mass experiments will be conducted in selected reactor assemblies to determine the influence of core density change on the critical mass of near-equilateral cylinders with hydrogeneous reflector.

The critical mass data are of importance in several respects:

- a. for application in resin columns, and
- b. for application of interaction criterion based on density formulation.

7. Limiting Concentration for Aqueous Pu Solution and Check on Value of  $\eta$

An experiment is considered to evaluate the limiting concentration of Pu in an aqueous solution from critical experiments. The limiting concentration is the smallest concentration which can be made critical in an "infinite" system. The value of  $\eta$  will also be determined from the experiment. A large sphere (somewhat < 42 inches in diameter) is being considered for the reactor vessel in this experiment.

8. Effect of Higher Isotopes of Plutonium  $\text{Pu}^{240}$ ,  $\text{Pu}^{241}$  on Criticality

In dilute aqueous plutonium solutions,  $\text{Pu}^{240}$  acts as a neutron poison; the effect of  $\text{Pu}^{240}$  on the critical mass will vary with plutonium concentration (H/Pu ratio), through dependence upon the neutron energy spectra. In predominantly fast systems,  $\text{Pu}^{240}$  assumes the role of a fissile isotope. On the other hand, high exposure plutonium, such as that obtained from power reactor fuels, will also contain non-negligible amounts of the highly fissile  $\text{Pu}^{241}$  isotope in addition to the  $\text{Pu}^{240}$ . Experiments will be conducted with both dilute and concentrated solutions containing these isotopes.

The criticality studies will include plutonium with a nominal  $\text{Pu}^{240}$  content ranging up to 15-20 percent; the experiments will also include several different intermediate isotopic contents. These measurements are subject to the availability of the high exposure plutonium.

9. Temperature Coefficients

Since plutonium has a large fission resonance cross section adjacent to the thermal region, the temperature coefficient for a plutonium system will differ from that of uranium. It is desirable to know this coefficient in connection with hazards evaluations, and for evaluating nuclear safety of those processes in which elevated temperatures may be encountered. Only unpressurized systems with temperatures below 100 C are being considered.

10. Soluble Poisons

Safety by geometry and batch limits are the most common safety principles observed in the processing of fissile materials. Control through the use of soluble poisons has also been considered, but dropped in some instances for lack of information. Thus,

information on the effect of soluble poisons on the criticality of various systems will be developed. The poisonous effect of the standard components in certain systems, such as the nitrogen in plutonium nitrate solutions, will also be studied.

11. Resonance Detector and Cadmium Ratio Measurements

In connection with the above reactor assemblies, resonance detector and cadmium ratio measurements will be performed to gain further insight into the role which epithermal absorption plays on the criticality of plutonium systems. Foil irradiations will be made in selected reactor assemblies to determine the spectral index (effective neutron temperature) for plutonium critical assemblies. A more detailed knowledge of neutron spectra is desired, since proper values of group-averaged spectrum-weighted cross sections are not available for calculational schemes.

12. Reactivity Coefficient Measurements

The effect upon the reactivity of the system of substituting various materials for a void or a fuel sample will yield a "reactivity coefficient". These measurements will be conducted in selected reactor assemblies; the results will be of use to future users of the data, both as known perturbations on the basic systems and as check points in theoretical treatment of the reactor assemblies. The data will also be of the value for evaluating the neutron worth of fixed poisons for use in criticality control.

13. Measurement of Prompt Neutron Lifetime and  $k_{\text{eff}}$  from Pulsed Neutron Source Experiments and from Pile Noise Analyzer

The prompt neutron lifetime is an important parameter in the study of reactor kinetics. Measurements will be made of the neutron lifetime in selected reactor assemblies. Consideration

will be given to the feasibility of using pulsed neutron source techniques for determining effective multiplication constants of subcritical units.

14. Minimal Critical Mass for Nonuniform Fuel Distribution

The minimal critical mass does not occur for a homogeneous uniform concentration of fuel, but rather for that distribution which results in a "flat flux". Theoretical predictions indicate a reduction of as much as 30 percent over a uniform fuel distribution.<sup>(23)</sup> Several experiments are planned to check these results for plutonium systems.

Heterogeneous Systems

1. Plutonium Metal-Plutonium Solution System; Plutonium-Alloy Fuel Elements in Water Lattices

The dissolution of plutonium metal in recovery operations, and the manufacture of plutonium-alloy fuel elements for the Plutonium Recycle Test Reactor are examples of a need for critical mass information pertaining to moderated heterogeneous plutonium systems. As with homogeneous systems, the effect on the critical mass of the H/Pu ratio, plutonium isotopic content, etc., will be studied. Plutonium dissolution is of special interest since in this case, the plutonium metal (scrap) will exist in a plutonium nitrate solution being moderated and reflected by this solution.

2. Plutonium Enriched Uranium Systems

It is anticipated that in connection with the reprocessing of nonproduction fuels from power reactors and the Plutonium Recycle Program, a need will arise for critical mass data on heterogeneous and homogeneous systems composed of natural or depleted uranium which has been enriched in Pu; the necessary critical mass experiments will be performed as required.

### Initial Experiments for the Laboratory

Current plans for the initial criticality experiments in the Plutonium Critical Mass Laboratory are given below. These plans, similar to the plans of any research and development proposal, are subject to change in the light of new information which will undoubtedly develop as the program progresses.

The following vessels have been chosen for use in the initial series of criticality experiments with plutonium nitrate solutions.

	<u>Diameters</u>
Cylinders (Bare)	11, 12, and 13 inches
Spheres (Bare and Reflected)	12, 13, and 14 inches

The first criticality experiments will be conducted with the spherical vessels. Types of reflectors being considered for the first spherical assembly are water and concrete with and without Cd sheet interposed between the reflector and the criticality vessel.

These measurements will be used as a normalizing point to the P-11 experiments and also to obtain needed data for the further development of theoretical methods of critical mass calculations.<sup>(6)</sup>

Following these measurements, a series of experiments will be conducted with cylinders (bare); the experiments will then be extended to include fully reflected cylindrical vessels. Additional spherical, and cylindrical, reactor vessels will be fabricated; the dimensions of these vessels will depend on the results of the initial measurements.

### Experiments with Precipitates of Plutonium or Facsimiles

A split-half machine is currently being designed to facilitate criticality studies with plutonium-oxide hydrogen-moderated mixtures, which because of the H/Pu atomic ratio, are not available in liquid form.

Criticality experiments will be conducted with  $\text{PuO}_2$ -plastic mixtures or other similar materials;<sup>(24)</sup> small blocks of various dimensions up to two inches on a side, which have been adequately sealed or jacketed for

contamination control will be used. The first experiments will consist of bare and reflected rectangular parallelepipeds (cubical units). The H/Pu atomic ratio of the material for the initial experiments is tentatively 15, with a nominal plutonium density of 1.8 g/cc. A series of experiments will be conducted which involve different H/Pu ratios.

The experiments will subsequently be extended to include cylindrical and slab geometry; in the case of cylindrical geometry, the critical assembly will be built up from thin disks or pancakes of plutonium plastic mixtures; both bare and reflected assemblies will be studied.

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